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13. ABSTRACT (Maximum 200 words)

This document contains the final report for above grant. It contains a description of the problems studied, a summary of the most important results, a bibliography and list of publications supported by this grant.

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FINAL PROGRESS REPORT

1. **CONTRACT OR GRANT NUMBER:** DAAH04-96-1-0028
2. **PERIOD COVERED BY REPORT:** April 1, 1996 - March 31, 2000
3. **TITLE OF PROPOSAL:** Ultrafast Phenomena in Semiconductor Nanostructures: Coherence versus Dissipation
4. **NAME OF INSTITUTION:** University of Illinois at Chicago
5. **AUTHORS OF REPORT:** Walter Poetz

PROJECT SUMMARY

A theoretical investigation of coherence versus phase breaking has lead to a microscopic understanding of optical and structural coherent control processes in semiconductor nanostructures. Three fundamentally different electromagnetic coherent control schemes have been subjected to a microscopic theoretical analysis. We have shown that, on time scales below characteristic phase breaking times, quantum interference of competing pathways significantly changes fundamental material properties of semiconductors, such as optical absorption, optical gain, and LO phonon emission times. In the quantum dynamics regime, the relative phase between two or more perturbations is a powerful tool to control inter(sub)band transitions on a subpicosecond time scale. In addition, extent and limitations of structural control of Fano resonances in semiconductor heterostructures has been studied numerically. Our results provide support to the idea that future generations of ultrafast optoelectronic semiconductor devices may indeed be based on quantum-coherent control schemes.

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I. PROBLEMS ADDRESSED WITHIN THIS GRANT

Coherent control uses the principle of quantum interference to manipulate the dynamics of a quantum system. The basic idea is contained in Fermi's golden rule (as well as Young's double slit experiment), which says that in the quantum regime (wave regime) the response of a system in the presence of two perturbations is, in general, not equal to the sum of the responses to the individual perturbations, *i. e.*,

$$|\langle i|H_1 + H_2|f\rangle|^2 \neq |\langle i|H_1|f\rangle|^2 + |\langle i|H_2|f\rangle|^2 \quad , \tag{1}$$

where Hamiltonian H_1 and H_2 account for two different perturbations which can take the system from state $|i\rangle$ to $|f\rangle$. Rather, there is an interference term which can be manipulated by adjusting the relative phase between H_1 and H_2.

In this project we have used this fundamental principle of quantum mechanics as a guide to conduct a theoretical study of feasibility and limitations of coherent control of inter(sub)band transitions in semiconductor heterostructures. The theoretical framework in form of Boltzmann–Bloch equations derived from non–equilibrium Green's functions is based on earlier work of ourselves which was supported by the U.S. Army Research Office, Grant DAAL03-92-G-0368.[1] This approach was extended to electronic intersubband transitions and the electron–phonon interaction in quantum wells. Extensions of this approach have allowed us a microscopic theoretical study of coherent dynamics and phase breaking in multi–(sub)band semiconductor systems.

Three elementary control schemes which we have explored are shown in Fig. 1.[2] Adopted from atomic physics, interference between single– and multi–photon absorption in Fig. 1(a) has been used to control the symmetry of final states and transition cross sections.[3] In semiconductors, this scheme has been used to control photocurrents.[4,5] Another scheme, Fig. 1(b) uses two coherent light fields of variable time–delay, polarization, and/or frequency detuning. Heberle and coworkers have used this type (with variable time–delay and relative polarizations, but equal frequencies) to coherently control exciton formation (optical absorption) and Faraday rotation.[6] Control scheme 1(c) represents a driven three–level system and has been used to demonstrate stimulated Raman adiabatic processes in atomic systems,[7]

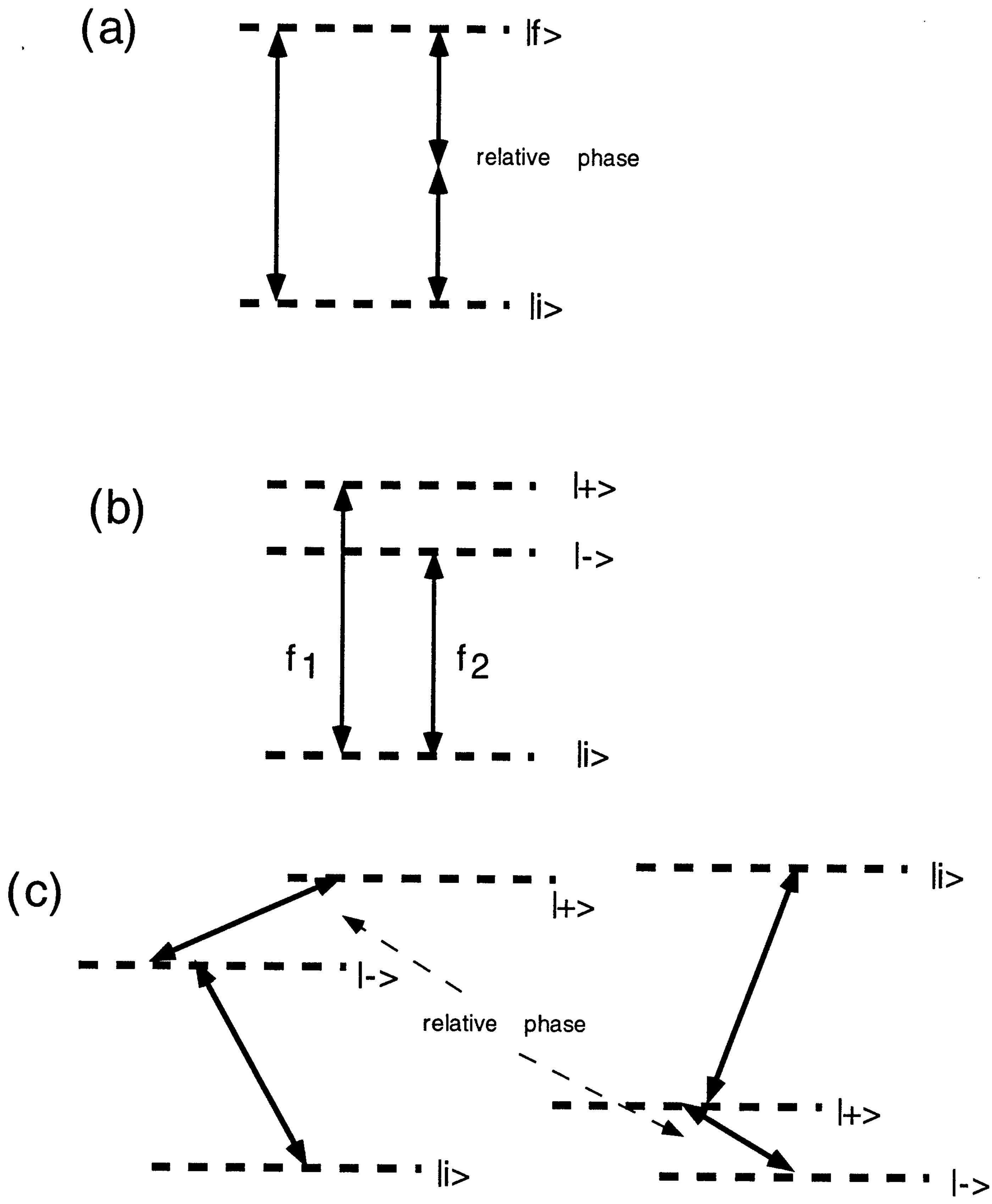

Fig. 1. Three control schemes investigated in within this grant. Dashed horizontal lines indicate the position of electron (sub)bands. Solid double-arrows indicate light fields. Note that scheme (c) exists in the V-configuration (left) and Lambda-configuration (right).

and is the basis for electromagnetically induced transparency demonstrated in various media.[8,9] The physical mechanism behind this scheme is that interband polarization f_{+-} between the doublet $+/-$ enters the kinetic equations for interband polarization between the doublet and the third subband (3), *i.e.*, $f_{\pm 3}$. The latter govern transitions between 3 and the doublet. Hence, when the Rabi period associated with the driven doublet is larger than a phase–matched pump (or probe) pulse coupling subband 3 to the subband doublet $+/-$, the phase of the driving field matters. In case of coherent control of LO phonon emission in interband transitions, the electron–phonon coupling plays the role of one of the competing perturbations ("the probe field"), making this an effect without counterpart in atomic systems.[10]

Another application of (1) uses structural engineering of semiconductor nanostructures to engineer Fano resonances.[11] These arise when there are two or more excitation pathways from a bound state to a continuum. Various structural design studies have been offered to evaluate the potential of semiconductors to exhibit Fano resonances in the absorption lineshape.[12–16] Fano lineshapes have been used in model studies to explore the possibility of manipulation of optical gain in semiconductors.[17]

The aim of our work was to develop a theoretical framework which allows us to design and evaluate coherent control schemes in semiconductors. Most of our studies based on manipulation by electromagnetic fields used asymmetric GaAlAs/GaAs–based heterostructures to provide the desired electronic structure. This heterostructure has a sufficiently large parameter range to allow the study of a variety of control phenomena. In particular, all three control schemes of Fig. 1 may be explored. Main difference to atomic systems is that (a) electronic levels are electron (sub)bands and (b) phase breaking times frequently lie in the sub–picosecond regime. GaAlAs/GaAs–based heterostructures of more general nature were used to structurally control Fano resonances in the electron intersubband photoabsorption spectrum.

II. SUMMARY OF MOST IMPORTANT RESULTS

A. Coherent Manipulation of Electron Inter(sub)band Transitions in Semiconductors using Electromagnetic Fields

(a) Coherent Control of Photoabsorption

All schemes represented in Fig. 1 were shown to allow coherent control of charge carrier absorption in semiconductors. Scheme 1(a) uses interference between single– and two–photon absorption to control the net photoabsorption cross section.[3–5] This effect was studied by us theoretically for semiconductor double wells.[18] Experimental means to demonstrate this effect are presently available. They include second–harmonics generation from a primary pulse with photon energy of approximately half of the main energy bandgap.

Scheme 1(b) was evaluated for two identical pump pulses, except for slightly detuned center frequencies (corresponding to typically 10–15 meV). It was shown to allow coherent control of photoabsorption provided that the duration of the two pump pulses does not exceed significantly the inverse beating frequency associated with the two pump fields. Both bulk semiconductors and double wells have been studied.[19,20] Again, present technology is available to study this effect experimentally.

For scheme 1(c) a coherent microwave field was chosen to resonantly couple an electron subband doublet in a double well and to provide interband coherence to manipulate absorption from a hole subband.[21] This scheme requires phase–locking to the pump pulse and high intensity (approx. 1 MW). To our knowledge such light fields are currently not available but would be highly desirable for a variety of other intersubband excitation studies.

We find that typically variations between 50 percent and 150 percent of single–pulse absorption can be achieved by variation of the relative phase between the two light fields. Theoretical limits based on the two–slit picture are zero and 200 percent. The subband nature, laser bandwidth, and many–body effects account for this difference.

(b) Control of Coherent Charge Oscillations in Asymmetric Semiconductor Double Wells

All schemes described in (a) applied to double wells allow control of coherent charge oscillations in asymmetric double wells.[21,18–20] This implies a coherent control of THz radiation

from these systems and may be used to either verify the effect or to generate THz pulses. In particular, the two–color pump scheme of Fig 1(b) in the long–time limit was investigated and shown to induce coherent charge oscillations.[2] However, in this case there is no phase sensitivity regarding the intensity of the THz signal.[2] We find that many–body effects in form of density–dependent band–gap renormalization, excitonic effects, and phse breaking (scattering) greatly limit the output intensity.

(c) Coherent Control of Final–State Population

Scheme 1(c) may be used to coherently control final state population as follows. Consider a pulsed phase–matched microwave field to arrive with the pump field. If the microwave field resonantly couples the upper doublet photoexcited electrons can tunnel between wide and narrow well.[21] The duration of the microwave field can be adjusted so as to either trap the carriers predominantly in the wide or the narrow gap. Hence, predominantly direct or indirect excitons may be generated.[22] Direct application of the STIRAP scheme was not found successful.[7] Phase breaking times are too short to allow coherent adiabatic dynamics to occur at moderate carrier densities ($\approx 10^{10}$ cm^{-2}).

(d) Coherent Control of Optical Gain in Electron Intersubband Transitions

We were able to demonstrate feasibility of optical gain without inversion for intersubband transitions in semiconductor heterostructures.[23] Scheme 1(c) was used where the doublet lies below the singlet subband. Again, a resonant microwave (mw) field was used to coherently drive the doublet. Depending on the relative phase between mw field and pump field net gain could be achieved in selected spectral regimes. Hence, we predict the possibility to coherently control optical gain. Calculations were performed both for incoherent pumping of the upper (third) subband and current injection. Results for both cases were quantitatively comparable, but somewhat more favorable for optical pumping as tunneling into a continuum provides an additional phase breaking agent.

(e) Coherent Control of LO Phonon Emission Rates

A completely new form of coherent control was predicted for a structure as discribed in (d) where, however, the role of the "probe field" was played by optical phonons. For this purpose the separation between doublet and upper singlet subband was chosen to be slightly

smaller than the relevant LO phonon energy of the structure. If a sufficiently short pump pulse is used to populate the uppermost subband, the scattering rate into the doublet via LO phonons is controlled by the relative phase between pump pulse and mw field. Variation of the relaxation time between 200 and 900 fs was predicted for a GaAs/AlGaAs-based double well. In terms of the simple 2–slit picture, this effect is based on quantum interference between single LO phonon emission and LO phonon emission combined with a mw photon emission and re–absorption.

(f) Coherent Control of Spin Polarization in Bulk Semiconductors

The two–color pump scheme of (a) (Fig. 1(b)) has been extended to circularly polarized pump pulses and has been shown to allow the generation of spin–polarized electron–hole pairs as a function of relative phase between the two pulses.[24]

B. Structural Control of Quantum Interference in GaAs/AlGaAs Heterostructures

Structural manipulation of quantum interference based on Fano resonances has been shown to allow control of optical absorption spectra of semiconductor heterostructures. We investigated a variety of structures, all based on readily available GaAs/AlGaAs material.[16,20] Structures have been investigated which show this effect for a single resonance, as well as two resonances. In the former case, the importance of overlap of the initial–state wave function with continuum scattering states was demonstrated. In the latter case, resonances must be closely spaced (typically 25 meV) to show a significant effect. Fano resonances may be controlled by electric fields in properly doped systems.[20]

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III. LIST OF SCIENTIFIC ARTICLES

(A) SUBMITTED:

W. Pötz, "Quantum coherent control of Coulomb enhancement by Two–Color Pulse Shaping", submitted March 2000.

(B) IN PRESS:

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W. Pötz, "Damping of Charge Oscillations in Double Wells: Macroscopic Manifestation of Phase Breaking in Semiconductors" (Lecture Notes), Research Workshop on Condensed Matter Physics, June 17 to August 23, 1996, ICTP, Trieste, Italy.

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(D) PhD Thesis:

- T. Kevosheeva, "*Coherent Control in Bulk Semiconductors by Multiple Optical Light Fields*" Ph.D. Thesis, supervised by W. Pötz, University of Illinois at Chicago, expected to be completed by June 2001.

IV. LIST OF PARTICIPATING SCIENTIFIC PERSONNEL AND DEGREES AWARDED

W. Pötz (PI, two summer months, full period)

Xuedong Hu, Postdoctoral Research Fellow, July 1996 – 1998 (2 years)

M. Pustilnik, Postdoctoral Research Fellow, Sept 1996 – Aug.1997 (1 year)

T. Kivosheeva, Ph.D. student (2 semester)

V. OTHER RELEVANT ACTIVITIES

PRESENTATIONS AT CONFERENCES AND WORKSHOPS

W. Pötz, "Optical and Structural Control of Quantum Interference in Semiconductors", US–Japan Workshop on the Quantum Control of Molecular Reaction Dynamics, Honolulu, Dec. 12–15, 1999 (invited).

W. Pötz, "Quantum Interference Phenomena in Electron Intersubband Transitions", 5-th Int. Conference on Intersubband Transitions in Quantum Wells, Bad Ischl, Austria, Sept. 7–11, 1999 (invited).

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W. Pötz,"Optical and Structural Manipulation of Quantum Interference in Semiconductor Nanostructures", Internat. Workshop on Novel Optical Materials, Jan. 11-12, 1999, College Station, Texas.(invited)

W. Pötz, "Optical Gain and Phonon Emission Rates in Semiconductor Quantum Wells ", "29th Winter Colloquium on the Physics of Quantum Electronics", Snowbird, Utah, Jan. 5–8, 1999 (invited).

W. Pötz, "Quantum Interference: Applications to Semiconductors and Devices?" Minisymposium on Nanostructure Electronics", Beckman Institute, Champaign–Urbana, Illinois, Nov. 13, 1998. (invited)

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CONFERENCE AND WORKSHOP ORGANIZATION

International Workshop on Coherent Control in Semiconductors, Chicago, May 19–22, 1998.

SPIE's International Symposium, Optoelectronics '99, member of program committee, January 1999, San Jose, CA.

Physics of Quantum Electronics, member of program committee ("Coherent Phenomena in Semiconductors"), January 1999, Snowbird, UT.

COLLOQUIA

"Optical and Structural Control of Quantum Interference in Semiconductor Nanostructures", Physics Department, University of Toronto, Canada, April 5, 1999.

"Quantum Interference and Coherent Control in Semiconductors", Physics Department, Northwestern University, Nov. 12, 1998.

"Young's Double Slit Experiment: Recent Applications in Atomic, Molecular, and Solid–State Physics", to students of SPS, UIC, Feb. 12, 1998.

"Coherent Control of Electronic Interband Transitions in Semiconductors", Feb. 6, 1998, Physics, University of Notre Dame.

"Manipulation of Carrier Dynamics and Optical Properties in Semiconductors", Armour College of Engineering and Science, Illinois Institute of Technology, Nov. 6, 1997.

"Coherent Control of Charge Oscillations and Light Absorption in Semiconductor Heterostructures, UIC Physics Condensed Matter Seminar, Nov. 96.

"Coherence versus Phase Breaking in Semiconductor Heterostructures", Univ. of Graz, Austria, Nov. 96.

"Coherence and Phase–Breaking in Semiconductor Nanostructures", Physics, University of Illinois at Chicago, April 17, 1996.

EDITORIAL WORK

Associate Editor for Applied Physics Letters (and Journal of Applied Physics)

Special Editor for Superlattices and Microstructures **26**(2), 1999.